

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE



In re Patent Application of	)	
Kari Harkonen et al.	)	Group Art Unit: 1762
Application No.: 10/642,426	)	Examiner: BRET P. CHEN
Filed: August 15, 2003	)	Confirmation No.: 9896
For: DEPOSITION OF CARBON- AND	)	
TRANSITION-METAL-CONTAINING	)	
THIN FILMS	)	

DECLARATION PURSUANT TO 37 C.F.R. § 1.132  
OF DR. MARKKU ANTERO LESKELÄ

Commissioner for Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450

Sir:

I, MARKKU ANTERO LESKELÄ, state as follows:

(1) I received M.Sc. degree in 1974 and a PhD in 1980 from Helsinki University of Technology. During 1974-80 I worked as a teaching assistant at Helsinki University of Technology, during 1980-86 as associate professor at the University of Oulu, and during 1986-1990 as a Professor at the University of Turku. Since 1990 I hold the position as Professor of Inorganic Chemistry at the University of Helsinki. My research activities have included thin films made by various chemical methods (Atomic Layer Deposition, electrodeposition and Successive Ionic Layer Adsorption and Reaction) for various applications in micro- and optoelectronics. Experience in ALD goes back to 1982. Another research area involves coordination compound catalysts and related metalorganic synthesis. The main emphasis in the catalyst research is in olefin polymerization and oxidation catalysts. I have published

420 peer reviewed papers, 50 reviews, over 60 conference proceedings papers, and hold several patents.

(2) I have devoted much of my career to the study of thin film and surface processes, including atomic layer deposition.

(3) I have studied the above-identified United States Patent Application, the Official Action dated March 7, 2006, the scientific reasoning expressed in the Official Action, and the contents of the four United States Patents discussed in the Official Action. The cited United States Patents were studied while focusing on what they teach to persons skilled in the ALD chemistry and technology.

(4) I have no association with this Patent Application, and am being compensated for my study time at my customary level for providing consultation service. I am not an employee, officer, director or shareholder of the owner of this Patent Application. I do not have any research project that is funded by the owner of this Patent Application.

(5) ALD is not a new technology, however, the ALD chemistry is not very developed and mature. The development has been very empirical and the theoretical work has so far been limited to modeling of the known processes. No new ALD reactions have been developed based on predictions or modeling. Thus, it can be said that ALD is recognized by those skilled in the art to require empirical testing following a consideration of the known chemistry of the respective source chemicals before a successful outcome can be appraised.

(6) In my opinion the Patent of Elers (U.S. Patent 6,482,262) is more limited in its teaching than as stated in the Official Action. The patentee did not indicate that any real organometallic chemical is suitable for use as the carbon

source chemical. The patentee indicates several times that the carbon source chemicals are compounds of B, Si, P, or a hydrocarbon, such as an alkane.

(7) True "organometallic" compounds are defined as compounds having direct metal-carbon bond. Technical journals and other publications, such as U.S. Patents 4,378,987; 4,442,888 and 5,933,760 sometimes refer to compounds of B, Si, and P containing organic groups as being "organometallic". However, it is clear for chemists that such compounds do not include a metal. This type of misnaming is tolerated in technical literature but the chemists recognize immediately that they are not true organometallics.

(8) The metals and non-metals, like B, Si, and P, and their compounds are recognized to have different properties and reactivities. This is confirmed by a look at the Periodic Table of Elements where a dark line is often drawn between metals and non-metals. See the copy of the "Periodic table of the elements" that is attached. Thus elements such as B, Si and P are located on a different side of this line than metals like Al. The chemists are well aware of the significance of this line.

(9) The differences in properties of metals and non-metals and their compounds are known by the chemists. Compared to non-metals like B, Si and P, metals such as Al are characterized not only by different physical properties but different chemical properties like lesser electronegativity and different oxidation states. When bound to organic ligands the metalorganic compounds are characterized by less chemical stability and participation of d-orbitals in bond formation.

(10) Boron is a special case. It belongs to the same group in the Periodic Table as aluminum. Boron is, however, very different from the other group

members: it is not a metal, it is clearly more electronegative than the others, and resembles closely carbon. Among main group elements the first members in each group obey diagonal similarity instead of group similarity.

(11) True organometallic compounds are less stable than the organic compounds of non-metals.

(12) In my opinion (i) the precise and limited teachings of U.S. Patent 6,482,262 with respect to the carbon source chemicals, (ii) absence of data showing that one could cross the dark line in the Periodic Table when choosing carbon precursors, and (iii) the recognized differences in the reactivity if one were to cross the line in the Periodic Table, would not enable one of ordinary skill in the ALD technology to conclude that there would be a successful outcome as claimed in this Patent Application in the absence of experimental research if the dark line in the Periodic Table were crossed. Such conclusion is contrary to that expressed at the bottom of page 3 of the Official Action.

I declare that all the statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true. These statements were made after having been informed that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that willful false statements may jeopardize the validity of the Application and any Patent issued thereon.

Date:

August 31, 2006



Markku Antero Leskelä

**Curriculum Vitae**  
**MARKKU ANTERO LESKELÄ**

Professor of Inorganic Chemistry (Sept. 1, 1990- present), Department of Chemistry, FIN-00014  
University of Helsinki, Finland



- Personal data Born: Nov. 25, 1950; Kokkola, Finland; Married; three children
- Education **Helsinki University of Technology** : Master of Science (Eng.) 1974, Licenciate of Technology (Eng.) 1976, Doctor of Technology 1980
- Professional **Helsinki University of Technology**: Teaching assistant (1973-79), Acting professor (1981-82, 1986) ; **University of Oulu**: Acting associate professor (1979-81), Associate professor (1982-86), Acting professor (1985); **University of Utrecht**: Visiting scientist (1983); **University of Turku**: Professor (1986-90); **University of Florida**: Visiting scientist (1987-88); **P&M Curie University (Paris)**: Visiting professor (May-June 1999); **Academy professor**, 2004-2009
- Research interests Thin films and surface processes; Polymerization and oxidation catalysts; Nanostructures and nanomaterials
- Professional Societies Finnish Chemical Society 1973 -; Finnish Technical Society 1973 - International Confederation for Thermal Analysis 1980 -; American Vacuum Society 1989 -; American Chemical Society 1994 -; Materials Research Society 1995 -
- Honors, Awards Elected member of Finnish Academy of Science and Letters 1991 - Elected member of Academy of Technical Sciences 1996 - Magnus Ehrnrooth Foundation Award in Chemistry 2002 SVR I, 2004 Elected member of Finnish Society of Sciences and Letters 2005 -
- Publications 420 publications in refereed journals (about 170 on ALD), 49 review papers, 9 books and edited books, 63 papers in conference proceedings, 385 abstracts of conference presentations, 23 patents/applications, 30 other publications
- Major professional activities Finnish Chemical Society: member of the board 1991 (vice chairman 93-94, chairman 95-96); Association of Finnish Chemical Societies: member of the board 1994 (vice chairman 97-98 ); Finnish Chemical Congress: program chairman of the Chemical Congress 1997-1999; International Confederation for Thermal Analysis: member of the council 1985-92 Finnish Academy of Science and Letters: secretary of the chemistry chapter 1994 2003, chairman 2004- ; Scientific Council of Chemical Industry Federation in Finland 1994 – 2000; Nordic Research Academy (NorFA) member of the scientific board for Science 1997- 2001; International Union of Pure and Applied Chemistry, Commission on High

Temperature Materials and Solid State Chemistry: associate member 1997 – 2001, Division of Inorganic Chemistry: titular member 2006 –

Referee reviewer for 9 professor positions Uppsala, Luleå, Oslo, Delft, Oxford, Lausanne, McMaster, Florida, Florida State), 9 docent positions, and 25 doctoral theses.  
Official opponent for 17 doctoral dissertations (incl. Oslo, Tallinn, Utrecht, Delft, Ghent)

Foundations Orion Research Foundation: member of the board 1992 – ; Foundation for Chemical Congress: member of the board 1996 - ; Gust Komppa Fund (A. Kordelin Foundation): chairman 2002 –; Outokumpu Foundation: member of the board 2003 –

Scientific journals Member of editorial board: Acta Chemica Scandinavica 1989 – 1999; Displays and Imaging International Edition 1997 –; Critical Reviews in Solid State Science 2004 -  
Regular referee in 12-15 journals; annually 20-35 papers referred

Researcher training Number of PhD students supervised: 36. Since 2000 Wei-Min Li 2000, Kari Mäkelä 2000, Heini Saloniemi 2000, Marika Juppo 2001, Tapio Kanninen 2001, Sari Paavola 2002, Antti Rahtu 2002, Marianna Kemell 2003, Nora Mäkelä-Vaarna 2003, Terhi Nissinen 2004, Mika Kettunen 2004, Raija Matero 2004, Kaisa Kervinen 2005, Petro Lahtinen 2005, Titta Aaltonen 2005, Kristian Lappalainen 2005, Pascal Castro 2005, Petra Alen 2005, Heikki Korpi 2005, Kirill Axenov 2005, Anna Moisala 2006  
  
Number of PhD students under supervision: 23  
Number of MSc thesis supervised: 150

Research projects since 2000 Atomic Layer Epitaxy growth of Al<sub>2</sub>O<sub>3</sub>, TiO<sub>2</sub>, SiO<sub>2</sub>, Cu, Pd and Mo on high surface area powders and as thin films (TEKES 1998-2001) FIM 1 890 000  
Solid state detectors and arrays for X-ray astronomy (ESA, Metorex Oy, 1998-2000) FIM 450 000  
Catalytic bleaching of cellulosa: synthesis of complexes (TEKES, 1998-2000) FIM 630 000  
Photovoltaics based on CuInSe<sub>2</sub> thin films (University of Helsinki, 1998-2000, IVO-foundation) FIM 700 000 + 35 000  
Characterization of volatile precursors for commercialization (TEKES, 1998-2000) FIM 1 160 000  
ALE-technology for thin film structures in opto- and microelectronics (TEKES, ASM Microchemistry Oy, Planar Systems Oy 1999-2001) FIM 2 000 000  
ESF-ALENET network (European Science Foundation, 1999-2002)  
Optical brighteners for paper industry (TEKES, 1999) FIM 143 000  
Fabrication of thin films for electronics by Atomic Layer Epitaxy and Electrodeposition (Academy of Finland, 1999-2002) FIM 1 910 000  
INTAS-project: Development of new single-component Pd(II)-based catalysts for alternating copolymerization of 1-olefins and CO (1999-2001)



Atomic Layer Deposition of high-k oxides for gate dielectrics (Motorola, 2000-2002, 2004-2005) USD 180 000 + 60 000

Metallocenes as polymerization catalysts (Academy of Finland, 2000-2002) FIM 1 100 000

New generation polymerization catalysts (Borealis Polymers Oy, 2000-03) FIM 2 000 000

New ALD processes for low refractive index materials (Planar Systems 2001-2002), 75 000 €

Nanochemical phenomena (TEKES, 2001-2004) 325 000 €

Metal complexes as selective initiators for ionic and radical polymerizations (Academy of Finland, 2001-2003) 100 000 €

Solution Deposition of thin films, COST 528 (2001-2005)

Bio- and nanopolymers Center of Excellence (Academy of Finland, TEKES, University of Helsinki, 2002-2007) 1 020 000 €

Fabrication of micro- and nanostructures by ALD and other surface chemistry controlled methods (Academy of Finland, 2002-2006) 150 000 €

New technologies for passive integration and ALD dielectrics for microelectronics (TEKES, ASM Microchemistry, 2002-2004) 225 000 €

Development of ALD of noble metal films (TEKES, ASMM, Environics Oy, 2002-2006) 244 000 €

Interactions between liquid phase and solid surfaces (TEKES Pinta-program, 2002-2006) 456 000 €

Control of small particle surface forces (TEKES Pinta-program, 2002-2006) 280 000 €

Oxidation catalysts for pulp bleaching (TEKES, Kemira Oyj, Stora-Enso Oyj, M-real Oyj, UPM-Kymmene Oyj, Andritz Oy, 2001-2004) 240 000 €

New coatings for medical instruments (Medtronic Ltd, 2002-2005) 100 000 €

TlBr hard X-ray imaging detectors (TEKES, Metorex Ltd, 2002-2004) 120 000 €

Low cost wiring method (Metso Oyj, Asperation Oy, Avantone Oy, 2003-2005) 60 000 €

Development of ALD processes for ferroelectrics and other microelectronics applications (ASM Microchemistry, 2004-2008) 1 000 000 €

Fabrication of nanomaterials and -structures via metalorganic synthesis and thin films (Academy of Finland, 2004-2009, Academy professor) 970 000 €

Development of ALD processes for  $\text{La}_2\text{O}_3$  (Honeywell Inc., 2005-2006) USD 120 000

1

Group  
IA

2

IIA

3

IIIA

4

IVB

5

VIA

6

VIB

7

VIIA

8

VIII

9

VIIIA

10

VIII

11

IB

12

IIB

13

IIIB

14

IVB

15

VB

16

VIB

17

VIIA

18

VIIIA

1

H

1.0079

3

Li

6.941

11

Na

22.9898

19

K

39.0983

37

Rb

85.4678

55

Cs

132.905

87

Fr

(223)

4

Be

9.01218

12

Mg

24.305

20

Ca

40.08

38

Sr

87.62

56

Ba

137.33

88

Ra

226.025

21

Sc

44.9559

29

Y

88.9059

39

Zr

91.224

57

La

138.906

89

Ac

227.028

22

Ti

47.88

40

Zr

91.224

72

Hf

178.49

104<sup>a</sup>

Unq

(261)

23

V

50.9415

41

Nb

92.9064

73

Ta

180.948

105<sup>a</sup>

Unp

(262)

24

Cr

51.996

42

Mo

95.94

74

W

183.85

106<sup>a</sup>

Unh

(263)

25

Mn

54.9380

43

Tc

(98)

75

Re

186.207

107<sup>a</sup>

Uns

(262)

26

Fe

55.847

44

Ru

101.07

76

Os

190.2

27

Co

58.9332

45

Rh

102.906

77

Ir

192.22

28

Ni

58.69

46

Pd

106.42

78

Pt

195.08

29

Cu

63.546

47

Ag

107.868

79

Au

196.967

30

Zn

65.39

48

Cd

112.41

80

Hg

200.59

31

Ga

69.72

49

In

114.82

81

Tl

204.383

32

Ge

72.59

50

Sn

118.71

82

Pb

207.2

33

As

74.9216

51

Sb

121.75

83

Bi

208.980

34

Se

78.96

52

Te

127.60

84

Po

(209)

35

Br

79.904

53

I

126.905

85

At

(210)

36

Kr

83.80

54

Xe

131.29

86

Rn

(222)

9

F

18.9984

17

Cl

35.453

35

Br

79.904

53

I

126.905

85

At

(210)

8

O

15.9994

16

S

32.06

34

Se

78.96

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Te

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Po

(209)

7

N

14.0067

15

P

30.9738

33

As

74.9216

51

Sb

121.75

83

Bi

208.980

6

C

12.011

14

Si

28.0855

32

Ge

72.59

50

Sn

118.71

82

Pb

207.2

5

B

10.81

13

Al

26.9815

31

Ga

69.72

49

In

114.82

81

Tl

204.383

2

He

4.00260

10

Ne

20.179

18

Ar

39.948

58

Ce

140.12

59

Pr

140.908

60

Nd

144.24

61

Pm

(145)

62

Sm

150.36

63

Eu

151.96

64

Gd

157.25

65

Tb

158.925

66

Dy

162.50

67

Ho

164.930

68

Er

167.26

69

Tm

168.934

70

Yb

173.04

71

Lu

174.967

90

Th

232.038

91

Pa

231.036

92

U

238.029

93

Np

237.048

94

Pu

(244)

95

Am

(243)

96

Cm

(247)

97

Bk

(247)

98

Cf

(251)

99

Es

(252)

100

Fm

(257)

101

Md

(258)

102

No

(259)

103

Lr

(260)

★ Lanthanide series

▲ Actinide series

Note: Atomic masses shown here are the 1983 IUPAC values (maximum of six significant figures). <sup>a</sup> Symbols based on IUPAC systematic names.

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Hawley's Condensed Chemical Dictionary,  
Eleventh Edition, Revised by N. Irving Sax  
and Richard J. Lewis, Sr., Van Nostrand,  
New York (1987)